

Experimental Test of Complementarity by Nuclear Magnetic Resonance Techniques

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Abstract

We have tested complementarity for the ensemble-averaged spin states of nuclei ^{13}C in the molecule of $^{13}\text{CHCl}_3$ by the use of the spin states of another nuclei ^1H as the path marker. It turns out that the wave-particle duality holds when one merely measures the probability density of quantum states, and that the wave- and particle-like behavior is simultaneously observed with the help of measuring populations and coherences in a single nuclear magnetic resonance(NMR) experiment. Effects of path-marking schemes and causes of the appearance and disappearance of the wave behavior are analyzed.

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Whether the wave-like and particle-like behavior, the fundamental attributes of quantum mechanical entities, can display or be observed simultaneously in a single experiment has been argued[1] since the foundation of quantum mechanics in 1920's. Bohr claimed the answer to be negative and expressed it as complementarity[2], one of the most basic principles of quantum mechanics. Bohr complementarity is usually illustrated by the famous two-slit experiment, in which a quantum object reaches a final state along two different paths simultaneously. The probability density or population of the final state displays interference when two paths are indistinguishable. Any attempt or practice to gain 'which-path' information unavoidably destroys the interference pattern. As the different path and the interference pattern are commonly attributed to a particle and a wave correspondingly, the results stated above are thus superficially referred to unobservableness of the particle- and wave-like behavior under the same experimental condition, in short, wave-particle duality of matter.

Complementarity was justified by analyzing the physical principles of early 'thought' experiments like Einstein's recoiling slit[1] or Feynman's light microscope[3] and recent experimental results with two trapped ions[4], an atom interferometer[5] and a pair of entangled photons[6] inspired by the proposal of Scully *et al*[7]. The mechanisms or origins that enforce complementarity, however, vary from one experimental situation to another[3,5,7-11]. In the 'thought' experiments with classical which-path markers, the act of labelling or measuring the paths invariably introduces a random momentum transfer or uncontrolled phase shift, thus washing out the interference fringes, which can be explained by Heisenberg uncertainty relation. While marking the path quantum-mechanically in the recent experiments does not lead to noticeable phase shift, disappearance of interference when measuring the probability density of the final state originates in quantum correlations of the which-path detector and observed quantum state, and the uncertainty principle plays no role.

In all previous variants of the two-slit experiment the wave behavior was observed by measuring the population in the final state, and the quantum state of the center-of-mass motion for micro-particles was investigated. Is complementarity applicable to the internal

states for a microscopic object or even a bulk ensemble? What are the effects of different path-marking schemes and the causes of appearance and disappearance of the wave behavior? Is there another way to detect the wave behavior of the quantum states except the population measurement? Most significantly, is observation of the wave and particle behavior under the same experimental condition absolutely prohibited indeed? These questions motivated us to perform this study by using an ensemble of nuclear spins as sample with NMR techniques.

Our experiment scheme is illustrated in Fig.1. An ensemble of molecules with two spin- $\frac{1}{2}$ nuclei is firstly prepared in the initial state $|\psi_i\rangle = |0\rangle_b |0\rangle_a \equiv |00\rangle$, where b is the observed nuclei, a is the labelling ones, and $|0\rangle$ and $|1\rangle$ represent the states with spin-up and spin-down respectively. In Fig.1a, an operation $R_i^b(\theta) = \begin{pmatrix} \alpha(\theta) & -\beta(\theta) \\ \beta(\theta) & \alpha(\theta) \end{pmatrix}$ on nuclei b transforms the initial state $|00\rangle$ into an intermediate state $|\psi_{1m}\rangle = [\alpha(\theta)|0\rangle_b + \beta(\theta)|1\rangle_b]|0\rangle_a$ (assuming $|\alpha(\theta)|^2 + |\beta(\theta)|^2 = 1$), which is then transferred to the final state $|\Psi_{1f}\rangle = \frac{1}{\sqrt{2}}[(\alpha(\theta) + \beta(\theta)e^{i\phi})|0\rangle_b + (\beta(\theta) - \alpha(\theta)e^{-i\phi})|1\rangle_b]|0\rangle_a$ by the operation $U^b(\phi) = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & e^{i\phi} \\ -e^{-i\phi} & 1 \end{pmatrix}$ on b . As the labelling nucleus a remains in the state $|0\rangle_a$ all the time as a spectator, two paths along which the nuclei b reaches the final state $|0\rangle_b(|1\rangle_b)$ through the intermediate state $|0\rangle_b$ and $|1\rangle_b$ are indistinguishable. The probability density of finding b in the final state $|0\rangle_b(|1\rangle_b)$ is measured to be $\frac{1}{2}[1 + 2\alpha(\theta)\beta(\theta)\cos\phi]$ ($\frac{1}{2}[1 - 2\alpha(\theta)\beta(\theta)\cos\phi]$). Repeating experiment by changing ϕ will produce Ramsey fringes denoted by the term $\pm\alpha(\theta)\beta(\theta)\cos\phi$, showing wave behavior obviously[12]. While in Fig.1b, the operations $R_2^{ba}(\theta) = CN_{ba} \bullet R_1^b(\theta)$ and $U^b(\phi)$ are successively executed, with CN_{ba} being the controlled-NOT gate in quantum computing, b and a , the control and target qubits respectively. The intermediate states $|\psi_{2m}\rangle = \alpha(\theta)|00\rangle + \beta(\theta)|11\rangle$ and the final state $|\psi_{2f}\rangle = \frac{1}{\sqrt{2}}\{|0\rangle_b [\alpha(\theta)|0\rangle_a + \beta(\theta)e^{i\phi}|1\rangle_a] + |1\rangle_b [-\alpha(\theta)e^{-i\phi}|0\rangle_a + \beta(\theta)|1\rangle_a]\}$ are thus obtained. Since the states $|0\rangle_b$ and $|1\rangle_b$ are definitely marked by $|0\rangle_a$ and $|1\rangle_a$ respectively in the preparation of the intermediate state, i.e., $|0\rangle_b \longleftrightarrow |0\rangle_a$ and $|1\rangle_b \longleftrightarrow |1\rangle_a$, and those markers remain unchanged in the later transformation $U^b(\phi)$, two paths along which spin b involves to the final state $|0\rangle_b(|1\rangle_b)$ from the intermediate states $|0\rangle_b$ and $|1\rangle_b$ simultaneously can be

precisely identified by the states of a . The probability densities of finding spin b in the final state $|0\rangle_b$ from the intermediate states $|0\rangle_b$ and $|1\rangle_b$ are $\frac{1}{2} |\alpha(\theta)|^2$ and $\frac{1}{2} |\beta(\theta)|^2$ respectively (the same for the final state $|1\rangle_b$). The particle behavior is thus clearly revealed. However, the probability density of finding b in the final state $|0\rangle_b$ or $|1\rangle_b$ is to be $1/2$, a constant independent on ϕ , due to the orthogonality of the quantum correlation states, so Ramsey fringes disappear. If one can detect the coherence, $\alpha(\theta)\beta(\theta)\sin\phi$, between the final states $|00\rangle$ and $|01\rangle$ (similar for $|10\rangle$ and $|11\rangle$) while measuring the probability density of those states, then the wave behavior reappears via the dependence of the coherence on ϕ , and the particle behavior is shown by the path marker and measurement of the population from different paths. Therefore, simultaneous observation of the wave and particle behavior in a single experiment is accomplished.

The sample we used is the molecule of carbon-13 labelled chloroform $^{13}\text{CHCl}_3$, nuclei ^{13}C and ^1H of which were chosen as observed nuclei b and labelling ones a respectively. Experiments were carried out on a Bruker ARX 500 spectrometer with conventional liquid NMR technique[13]. The resonance frequencies are about 125 MHz for ^{13}C and 500 MHz for ^1H , and the scalar coupling constant J_{ab} is near 215 Hz. Four lines, two for ^{13}C and two for ^1H spectra, can be well resolved. By applying two simultaneous line-selective pulses with appropriate frequencies and rotation angles as well as a magnetic field gradient pulse consecutively on the ensemble of nuclei ^{13}C and ^1H in thermal equilibrium[14], we prepared the quantum ensemble in an effective pure state expressed by the density matrix $\rho_{00}^i = AE + B|00\rangle\langle 00|$, where E is a 4×4 unit matrix, A and B are constants with $A \gg B$. As ρ_{00}^i has the same quantum-mechanical properties and NMR experimental results as the pure state $|00\rangle$ [15,16], the conclusions extracted from experiments on ρ_{00}^i can be applied to characterize $|00\rangle$. Operation $R_1^b(\theta)$ was implemented by a pulse $(\theta)_{-y}^b$, a pulse applied on ^{13}C and rotating a angle θ along the negative y-axis. The amplitudes of the wave function of the intermediate state thus obtained are $\alpha(\theta) = \cos(\theta/2)$ and $\beta(\theta) = \sin(\theta/2)$. Two values of θ were set at 90° and 53.24° in different runs, corresponding to the ratio $|\alpha(\theta)|^2 / |\beta(\theta)|^2$ of populations in two intermediate states to be 1 and 4. The controlled-

NOT gate CN_{ba} was achieved by a pulse sequence $(\pi/2)_{-y}^a(1/2J_{ab})(\pi/2)_y^{a,b}(\pi/2)_{-x}^{a,b}(\pi/2)_{-y}^b$, where $1/2J_{ab}$ is the time interval of free evolution period[17]. Transformation $U^b(\phi)$ could be denoted by $e^{-iI_x^b\theta_1}e^{-iI_y^b\theta_2}e^{-iI_x^b\theta_1}$ with $I_{x,y}^b$ being the spin operators of the nuclei ^{13}C and $\theta_1 = \tan^{-1}(-\sin\phi)$, $\theta_2 = 2\sin^{-1}(-\cos\phi/\sqrt{2})$, so it was performed by a pulse sequence $(\theta_1)_x^b(\theta_2)_y^b(\theta_1)_x^b$. Various values of ϕ were obtained by assuming appropriate θ_1 and θ_2 . So the wave and particle behavior in given situation could be examined by inspecting the dependence of populations and coherences on ϕ . Reading-out pulses $(\pi/2)_y^a$ and $(\pi/2)_y^b$ were finally applied and spectra for ^1H and ^{13}C were recorded. As the population difference and coherence of relevant states were transferred to the measured transverse magnetization after applying read-out pulses and embodied in the real and imaginary parts of the line intensity of the recorded spectra respectively[13], the normalized population and coherence of the interested states were extracted in Figs.2-4 by data analyzing and fitting.

Variations of the normalized populations $\rho_{00}^{f_1}$ and $\rho_{10}^{f_1}$ of the states $|00\rangle$ and $|10\rangle$ versus ϕ when two paths along which the spin ensemble of nuclei ^{13}C involve from the intermediate states to the final states cannot be discerned by the invariable state $|0\rangle_a$ of the spectator nuclei ^1H are shown in Fig.2. The range of ϕ is assumed in $(0, 2\pi)$ due to its 2π periodicity. Ramsey fringes can be clearly seen for two sets of data in the case of the ratio of the normalized populations in two intermediate states to be 1 and 4. The visibility of fringes decreases with the population ratio increased. These experimental results are in good agreement with the theoretical expectation. Experimental errors are mainly due to the inhomogeneity of the RF field and static magnetic field of the spectrometer, imperfect calibration of applied pulses (esp., the θ_1 and θ_2 setting in different runs), and signal decay during the experiment[17].

Variations of normalized populations versus ϕ when two paths from the intermediate to final states are distinguishable by the states of the labelling nuclei ^1H are depicted in Fig.3. In this case the populations, $\rho_0^{f_2}$ and $\rho_1^{f_2}$, of the observed nuclei ^{13}C in the final states $|0\rangle_b$ and $|1\rangle_b$ respectively don't vary with ϕ regularly, i.e., the wave behavior does not manifest itself in this way. Experimental data are also fairly consistent with the theory.

It can be concluded from Figs.2 and 3 that the conventional description of Bohr complementarity still holds for the quantum entity characterized by the internal states of a spin ensemble. The wave behavior displays in the measured populations when both paths are indistinguishable, and disappears in population measurements if one attempts to get which-path information. Nevertheless, we have measured the coherences, C_0 and C_1 , between the relevant state pairs ($|00\rangle, |01\rangle$) and ($|10\rangle, |11\rangle$) respectively, besides the population measurements when both paths were labelled by the states of nuclei ^1H . The measured coherences of the final states $|0\rangle_b$ and $|1\rangle_b$ (see Fig.4) also showed oscillatory property with ϕ , thus revealing the wave behavior in another way. Because the results given in Figs.3 and 4 were observed in the same time and under the same experimental situation, we then conclude that simultaneous observation of the wave and particle behavior of a quantum entity has been achieved in a single experiment.

Some remarks will be made as follows. In all previous two-slit experiments the wave behavior was solely observed through the population measurement. The results extracted from those experiments, including ours shown in Figs.2 and 3, confirmed that the oscillatory feature of populations excludes with the path distinguishability. Complementarity stated as "simultaneous observation of the wave and particle behavior is prohibited" is therefore valid in and only in the sense that one merely observes the population in his experiment.

As well known, observation of the wave behavior in classical and quantum physics requires that the relative phase between relevant wave components keep constant. The wave behavior can be manifested through the existence of coherence and population oscillation. As labelling different evolution paths by means of appropriate quantum states does not bring about any random phase shift and makes the phase difference between the wave function of the states involved unchanged, the wave character maintains after path-labelling. Although the populations in the final states remain invariable with ϕ , the oscillating coherence between states originated from different intermediate states and marked by labelling nuclei ^1H can surely be used to show the wave character. Therefore, realization of simultaneous observation of the wave and particle behavior of any quantum object with the help of quantum path

marking and measuring populations and coherence is physically reasonable and universal in principle. This scheme is readily feasible for the internal state of quantum systems using the techniques existing nowadays or available in the near future, e.g., for the internal state of atoms and molecules by optical spectroscopy. If some means of observing the coherence or other observable characterizing the wave behavior of the center-of-mass states of any quantum entity could be found out and executed in practice, simultaneous observation of the wave and particle behavior is then applicable to all quantum systems.

Besides the wave-particle duality, complementarity sometimes has endowed with more general concept. Two observable are referred to be complementarity if precise knowledge of one of them implies that all possible outcomes of measuring the other one are equally possible[7]. All canonical conjugate pairs (e.g., position and momentum) and two incommutable observable (e.g., longitudinal and transverse polarizations) are such examples. Complementarity in this general sense is consistent with current framework of quantum mechanics but beyond the scope of the present paper.

In conclusion, we have experimentally tested Bohr complementarity for the quantum entity characterized by the internal states of a nuclear spin ensemble in the bulk matter using concepts of the controlled-NOT gate and entangled states in quantum computing as well as techniques in NMR spectroscopy. Simultaneous observation of the wave and particle-like behavior of the quantum object was realized for the first time. The reasons why both behaviors could not be observed in the same time were revealed. The condition and method of observing the both in a single experiment were proposed. It was indicated that the method proposed in the present paper could be applied to the internal and center-of-mass states of a single micro-particle and macroscopic ensemble and would be nearly feasible for the internal state of some other quantum objects.

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Captions of the figures

Fig.1 Schematic of a 'two-slit' experiment with internal states of a quantum entity. The entity involves from the initial state $|00\rangle$ via the intermediate to final one by transformations of (a) $R_1^b(\theta)$ and $U^b(\phi)$; (b) $R_2^{ab}(\theta)$ and $U^b(\phi)$ (see text).

Fig.2 Normalized population vs. the relative phase ϕ when evolution paths are indistinguishable. Experimental data points \bigcirc and ∇ denote the populations $\rho_{00}^{f_1}$ in the state $|00\rangle$, $*$ and $+$ denote $\rho_{10}^{f_1}$ in the state $|10\rangle$. Theoretical curves expressed by $\frac{1}{2}(1 \pm \sin \theta \sin \phi)$ are depicted with the solid lines. (a) $\theta = 90^\circ$ and (b) $\theta = 53.24^\circ$.

Fig.3 Normalized populations vs. the relative phase ϕ when two paths are distinguishable. Data points \bigcirc , $*$, ∇ and $+$ denote $\rho_{00}^{f_2}$, $\rho_{01}^{f_2}$, $\rho_{10}^{f_2}$ and $\rho_{11}^{f_2}$, the notations of square and star denote $\rho_0^{f_2}$ and $\rho_1^{f_2}$ respectively. Theoretical curves expressed by $\rho_{00}^{f_2} = \rho_{10}^{f_2} = \frac{1}{2} \cos^2 \frac{\theta}{2}$, $\rho_{01}^{f_2} = \rho_{11}^{f_2} = \frac{1}{2} \sin^2 \frac{\theta}{2}$ and $\rho_0^{f_2} = \rho_{00}^{f_2} + \rho_{01}^{f_2} = \rho_1^{f_2} = \rho_{10}^{f_2} + \rho_{11}^{f_2} = \frac{1}{2}$ are depicted with the solid lines. (a) and (b) $\theta = 90^\circ$, (c) and (d) $\theta = 53.24^\circ$.

Fig.4 Coherence vs. ϕ when two paths are distinguishable. Data points \bigcirc and ∇ denote the coherence between the states $|00\rangle$ and $|01\rangle$, $*$ and $+$, between the states $|10\rangle$ and $|11\rangle$. Theoretical curves expressed by $\pm \frac{1}{2} \sin \theta \sin \phi$ are depicted with the solid lines. (a) $\theta = 90^\circ$, (b) $\theta = 53.24^\circ$.